Translation of selected passage from JP-A-081166/2001

Page 1, lower column:

(54) [Title of the Invention] Biodegradable Polyester Copolymer and Production Process Therefor

(57) [Abstract]

[Object] To provide a biodegradable polyester copolymer and a production process therefor, wherein the biodegradable polyester copolymer has superior processability as well as superior biodegradability and further is good in properties such as strength and extensibility.

[Means for solution] The biodegradable polyester copolymer is obtained by performing a transesterification reaction between polyethylene terephthalate and a dicarboxylic anhydride/ethylene oxide copolymer having an ethylene oxide content in the range of 50 to 100 mol % both exclusive. Favorable as an industrial production process is a process comprising the steps of the transesterification reaction and the extrusion molding of the resultant copolymer which are continuously performed by causing an extruder to extrude a mixture containing the polyethylene terephthalate, the dicarboxylic anhydride/ethylene oxide copolymer, and a catalyst while causing the extruder to heat and knead the mixture.

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* NOTICES *

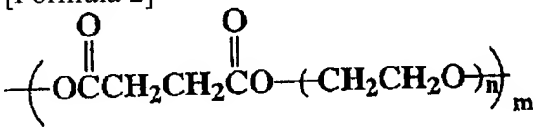
1.This document has been translated by computer. So the translation may not reflect the original precisely.
2.**** shows the word which can not be translated.
3.In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[The technical field to which invention belongs] While this invention is excellent in processability, properties, such as intensity and elongation, are related with a good biodegradability polyester copolymer and its manufacture method.
[0002]
[Description of the Prior Art] Although various synthetic-resin material is used in more various fields than before, while will remain as it is semipermanently, and the request of the environmental preservation in an earth scale in recent years will increase since the environmental load is very large if most of these synthetic resin does not have biodegradability but it is discarded in environment, it is just going to request strongly that a resin with biodegradability also as these resin material should be used as much as possible.
[0003] A duty of recycling-ization of a representing [from a rise of the request of such environmental preservation, / container recycling law is enacted recently and / by the PET (polyethylene terephthalate) bottle etc.]-on the other hand, polyethylene terephthalate is just going to be imposed. However, by the time most of the recycling to other uses is materialized difficult in the present condition by the recycling to a bevel-use PET bottle,, it will not result, but the present condition is that recycling-ization of a polyethylene terephthalate is not progressing to the degree expected in this way. For example, in a household-articles manufacture field, although it considers and gropes for recycling use of a disposal polyethylene terephthalate, the present condition is there being a problem to which processability's falls physical properties', such as intensity's and elongation's, falling, and having not resulted in practical use level.
[0004]
[Problem(s) to be Solved by the Invention] This invention aims at excelling in processability and offering a biodegradability polyester copolymer with a good property and its manufacture methods, such as intensity and elongation, while it is made in view of the starting technical background and is excellent in biodegradability.
[0005]
[Means for Solving the Problem] In order to attain the above-mentioned purpose, the polyester copolymer obtained by performing an ester exchange reaction between a polyethylene terephthalate, and an anhydrous dicarboxylic acid / ethylene oxide copolymer as a result of research comes to find out having excelled in processability and having the good property in intensity, elongation, etc., while excelling in biodegradability wholeheartedly, and this invention persons complete this invention.
[0006] That is, the biodegradability polyester copolymer concerning this invention is a polyester copolymer obtained by performing an ester exchange reaction between a polyethylene terephthalate, and the anhydrous dicarboxylic acid / ethylene oxide copolymer which has an ethylene oxide content in the range below 100 mol % exceeding 50-mol %. rubber since it excels also in processability, and properties, such as intensity and shock resistance, are also good and it has the ether chain of ethylene

oxide further, while the polyester copolymer pass such an ester exchange reaction is excellent in biodegradability -- a character [like] is presented and sufficient elongation is also provided
[0007] An anhydrous dicarboxylic acid / ethylene oxide copolymer is the following general formula (I).;

[Formula 2]



… (I)

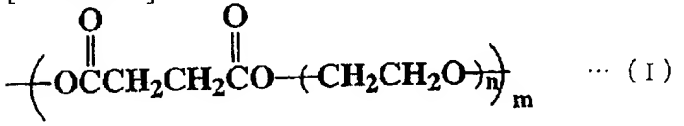
It is desirable that they are the succinic anhydride / ethylene oxide copolymer expressed with (however, the number exceeding 1 is shown by the inside n of a formula). While being able to improve such composition, then biodegradability further, there is an advantage which can improve further.
[0008] The ethylene oxide content in an anhydrous dicarboxylic acid / ethylene oxide copolymer can secure more excellent biodegradability and elongation, securing sufficient intensity with [thing / it is desirable that it is the 51-80 mol range of %, and] such a range.
[0009] As for the mixed weight ratio in the case of an ester exchange reaction, it is desirable that it is the range of an anhydrous dicarboxylic acid / ethylene oxide copolymer 20 - the 800 weight sections to the polyethylene-terephthalate 100 weight section. Sufficient intensity and sufficient elongation are securable, fully securing the biodegradability excellent in considering as such a range.
[0010] Moreover, the manufacture method of the biodegradability polyester copolymer this invention is characterized by performing extrusion molding of this biodegradability polyester copolymer while it makes an ester exchange reaction perform between a polyethylene terephthalate, and an anhydrous dicarboxylic acid / ethylene oxide copolymer and compounds a biodegradability polyester copolymer by carrying out heating kneading extrusion of the mixture containing the anhydrous dicarboxylic acid / ethylene oxide copolymer, and the catalyst which have a polyethylene terephthalate and an ethylene oxide content in the range below 100 mol % exceeding 50-mol % with an extruder. Since the generation of a copolymer and the fabrication of this copolymer by the ester exchange reaction can be performed continuously and the process to fabrication can be performed very efficiently by the heating kneading extrusion by such extruder, it is suitable as a industrial process.
[0011] As for an extruder, it is desirable that it is a biaxial extruder, and it can improve the manufacture stability in generation of the copolymer by the ester exchange reaction by this, and can manufacture the thing of the stable quality without dispersion continuously.
[0012] That it is 150 - 600 revolution per minute is the point whose much more efficient manufacture can make generate the copolymer by the ester exchange reaction more for a short time, and is attained, and the screw speed of an extruder has it. [desirable]
[0013]
[Embodiments of the Invention] The biodegradability polyester copolymer of this invention is a polyester copolymer obtained by performing an ester exchange reaction between a polyethylene terephthalate, and the anhydrous dicarboxylic acid / ethylene oxide copolymer which has an ethylene oxide content in the range below 100 mol % exceeding 50-mol %. The polyester copolymer obtained by performing an ester exchange reaction between such an anhydrous dicarboxylic acid / an ethylene oxide copolymer is excellent in processability, and, moreover, its properties, such as intensity, are good while it is excellent in biodegradability. Furthermore, since the ethylene oxide content is over 50-mol %, it has the ether chain of ethylene oxide in structure, and when the ether chain of such ethylene oxide exists, biodegradability improves notably. and rubber since it has the ether chain of ethylene oxide -- a character [like] is presented, and sufficient elongation is also provided, therefore it can use as a film, a sheet object, a Plastic solid, etc. in various uses
[0014] Especially as a polyethylene terephthalate, although not limited, it is desirable to use used polyethylene terephthalates, such as an abandonment PET bottle. Since recycling-ization of a polyethylene terephthalate can be attained by using such a used polyethylene terephthalate, it can fully

respond also to the request of environmental preservation.

[0015] Although especially the number average molecular weight of the polyethylene terephthalate to be used is not limited, the thing of the range of 10000-30000 is desirable.

[0016] Especially as an anhydrous dicarboxylic acid / an ethylene oxide copolymer, although not limited, it is the following general formula (I).;

[Formula 3]



... (I)

It is desirable to use the succinic anhydride / ethylene oxide copolymer expressed with (however, the number exceeding 1 is shown by the inside n of a formula). While being able to improve biodegradability further by considering as such composition, there is an advantage which can improve further.

[0017] Sufficient elongation and sufficient biodegradability can be made to have, the inside n of a formula of the aforementioned general formula (I) raising intensity more exceeding 1 with [thing / it is desirable that it is five or less range, and] such a range. Since it becomes impossible to secure sufficient intensity when n exceeds 5, it is not desirable. Especially, it is more desirable that it is the range of 2-5 the inside n of a formula of a general formula (I). In addition, when n is 1 (i.e., when it is the structure where it does not have an ether chain (when deviating from the range of this invention)), elongation will not fully be acquired, and biodegradability will also fall.

[0018] As for the ethylene oxide content in the aforementioned anhydrous dicarboxylic acid / ethylene oxide copolymer, it is desirable that it is the 51-80-mol range of %. Since intensity will fall if 80-mol % is exceeded preferably or on the other hand, since elongation sufficient less than [51 mol %] is no longer acquired upwards and biodegradability also falls, it is not desirable.

[0019] Although especially the number average molecular weight of the aforementioned anhydrous dicarboxylic acid / ethylene oxide copolymer is not limited, the thing of the range of 2000-20000 is desirable.

[0020] Moreover, as for the mixed weight ratio in the case of an ester exchange reaction, it is desirable to consider as the range of an anhydrous dicarboxylic acid / ethylene oxide copolymer 20 - the 800 weight sections to the polyethylene-terephthalate 100 weight section. In under 20 weight sections, since biodegradability falls upwards, and intensity will fall if the 800 weight sections are exceeded preferably or on the other hand, since sufficient elongation is no longer acquired, it is not desirable.

[0021] In addition, as for the number average molecular weight of the biodegradability polyester copolymer of this invention, it is desirable that it is 10000-50000. Since it is in the inclination for processability to fall when 50000 is exceeded preferably or on the other hand, since it is in the inclination for intensity to no longer be obtained fully, it is not desirable at less than 10000.

[0022] The biodegradability polyester copolymer concerning this invention can mix in a container a polyethylene terephthalate, the anhydrous dicarboxylic acid / ethylene oxide copolymer which has an ethylene oxide content in the range below 100 mol % exceeding 50-mol %, and a catalyst, is in a heating melting state, and can be stirred predetermined time under reduced pressure, and can be obtained by advancing an ester exchange reaction. Although the above-mentioned catalyst is not indispensable, it is desirable to use a reaction from a viewpoint advanced efficiently.

[0023] As a catalyst, especially if it functions as an ester exchange reaction catalyst, it will not be limited, for example, the tetramer object of titanium (IV) tetrapod isopropoxide, the monomer object of titanium (IV) isopropoxide, aluminum isopropoxide, tin octoate, etc. will be mentioned. It is desirable to use the tetramer object of titanium (IV) tetrapod isopropoxide and the monomer object of titanium (IV) isopropoxide also in these at a point excellent in especially the catalysis of the ester exchange reaction between a polyethylene terephthalate, and an anhydrous dicarboxylic acid / ethylene oxide copolymer.

[0024] Especially, as a industrial process, the following manufacture methods are suitable. That is, while

making an ester exchange reaction perform between a polyethylene terephthalate, and an anhydrous dicarboxylic acid / ethylene oxide copolymer and compounding a biodegradability polyester copolymer by carrying out heating kneading extrusion of the mixture containing the anhydrous dicarboxylic acid / ethylene oxide copolymer, and the catalyst which have a polyethylene terephthalate and an ethylene oxide content in the range below 100 mol % exceeding 50-mol % with an extruder, extrusion molding of this biodegradability polyester copolymer is performed.

[0025] There is an advantage which can perform continuously the generation of a copolymer and the fabrication of this copolymer by the ester exchange reaction by the heating kneading extrusion by such extruder. Moreover, since raw material mixture is kneaded by torque with the strong screw of an extruder, an ester exchange reaction can fully be advanced for a short time. furthermore, the temperature under kneading -- precision -- since it is held highly at predetermined temperature, repeatability is good, and there is no dispersion in the quality of the biodegradability polyester copolymer obtained, and it excels in manufacture stability

[0026] The above-mentioned heating kneading is usually performed under reduced pressure, and, as for the **, it is desirable to be referred to as 0.1 - 1Torr.

[0027] As for the heating temperature in the case of the above-mentioned heating kneading extrusion, it is desirable to consider as 260-320 degrees C. Since melting of a polyethylene terephthalate becomes inadequate at less than 260 degrees C and going on [of a reaction] becomes inadequate, if it exceeds 320 degrees C preferably or on the other hand, since it will become easy to produce dispersion in the quality of the biodegradability polyester copolymer obtained and manufacture stability will fall to it, it is not desirable. Especially, as for heating temperature, it is more desirable to consider as 280-290 degrees C.

[0028] The well-known extruder currently used for extrusion molding of a resin etc. general-purpose in the field of macromolecule processing as the above-mentioned extruder can be used. Usually, although a 1 shaft extruder or a twin screw extruder is used, it is desirable to use a twin screw extruder especially, dispersion is not produced in the quality of the biodegradability polyester copolymer obtained in this case, and manufacture stability can be improved further. In addition, although the extruder of a triaxial not less can also be used, since it becomes the low special type of versatility, it is expensive, therefore since a manufacturing cost is increased, it is not desirable.

[0029] Especially as a configuration of the screw of an extruder, although not limited, full flight type, dull MEJI type, mixing pin type, uni-melt type, a barrier form, etc. are mentioned, for example, and the thing of full flight type is used suitably especially.

[0030] Moreover, as for the rotational frequency of the screw of an extruder, it is desirable to set it as 150 - 600 revolution per minute. In less than 150 revolutions per minute, since time, i.e., reaction time, for the aforementioned mixture to pile up in an extruder increases, if 600 revolutions per minute are exceeded preferably or on the other hand, although the residence time is shortened, since it becomes easy to produce dispersion in the quality of the biodegradability polyester copolymer obtained, it is not desirable.

[0031] Moreover, as for ratio ratio of length to diameter of screw length (L) and the diameter of a screw (D), it is desirable to consider as the range of 30-90. Since the time required by extrusion will become long if 90 is exceeded preferably or on the other hand, since there is a bird clapper that less than 30 are insufficient as for an ester exchange reaction, it is not desirable. Especially, as for ratio of length to diameter, it is more desirable to consider as the range of 40-80.

[0032]

[Example] Next, the concrete example of this invention is explained.

[0033] The product made from <Material-of-construction> (polyethylene terephthalate A) Kanebo Synthetic fiber "bell pet EFG6C" (tradename), By the copolymer compounded by ring breakage copolymerization of a thing (succinic-anhydride / ethylene oxide copolymer X) succinic anhydride (SA) and ethylene oxide (EO) intrinsic-viscosity 0.7 dL/g and whose number average molecular weight are 20300 By the copolymer compounded by ring breakage copolymerization of a thing (succinic-anhydride / ethylene oxide copolymer Y) succinic anhydride (SA) and ethylene oxide (EO) whose

number average molecular weight composition ratios are SA/EO=49 / 51 (mol %), and is 7000 By the copolymer compounded by ring breakage copolymerization of a thing (succinic-anhydride / ethylene oxide copolymer Z) succinic anhydride (SA) and ethylene oxide (EO) whose number average molecular weight composition ratios are SA/EO=47 / 53 (mol %), and is 6500 By the copolymer compounded by ring breakage copolymerization of a thing (succinic-anhydride / ethylene oxide copolymer V) succinic anhydride (SA) and ethylene oxide (EO) whose number average molecular weight composition ratios are SA/EO=45 / 55 (mol %), and is 6000 By the copolymer compounded by ring breakage copolymerization of a thing (succinic-anhydride / ethylene oxide copolymer W) succinic anhydride (SA) and ethylene oxide (EO) whose number average molecular weight composition ratios are SA/EO=40 / 60 (mol %), and is 4500 That whose number average molecular weight composition ratios are SA/EO=21 / 79 (mol %), and is 2800.

[0034] after having put into the flask the mixture which made the <example 1> above-mentioned polyethylene terephthalate A, and the succinic anhydride / ethylene oxide copolymer X mix by the weight ratios 50/50, and put the tetramer object of titanium (IV) tetrapod isopropoxide into a flask as a catalyst subsequently so that it may become the 0.5 weight section to the aforementioned mixture 100 weight section, the biodegradability polyester copolymer be obtained by carry out a reaction (ester exchange reaction) at 290 degrees C under reduced pressure conditions for 30 minutes. In addition, the film created by the solvent cast method using this biodegradability polyester copolymer was transparent.

[0035] As a <example 2> catalyst, it replaced with the tetramer object of titanium (IV) tetrapod isopropoxide, and the biodegradability polyester copolymer was obtained like the example 1 except having used the monomer object of titanium (IV) isopropoxide.

[0036] The biodegradability polyester copolymer was obtained like the example 1 except having used the mixture which made the <example 3> polyethylene terephthalate A, and the succinic anhydride / ethylene oxide copolymer X mix by the weight ratios 70/30.

[0037] The biodegradability polyester copolymer was obtained like the example 1 except having used the mixture which made the <example 4> polyethylene terephthalate A, and the succinic anhydride / ethylene oxide copolymer X mix by the weight ratios 60/40.

[0038] The biodegradability polyester copolymer was obtained like the example 1 except having used the mixture which made the <example 5> polyethylene terephthalate A, and the succinic anhydride / ethylene oxide copolymer X mix by the weight ratios 80/20.

[0039] The biodegradability polyester copolymer was obtained like the example 1 except having used the mixture which made the <example 6> polyethylene terephthalate A, and the succinic anhydride / ethylene oxide copolymer X mix by the weight ratios 40/60.

[0040] The biodegradability polyester copolymer was obtained like the example 1 except having used the mixture which made the <example 7> polyethylene terephthalate A, and the succinic anhydride / ethylene oxide copolymer X mix by the weight ratios 20/80.

[0041] It replaced with the <example 8> succinic anhydride / ethylene oxide copolymer X, and the biodegradability polyester copolymer was obtained like the example 1 except having made the catalyst addition into the 0.2 weight section to the mixture 100 weight section, using a succinic anhydride / ethylene oxide copolymer Y.

[0042] It replaced with the <example 9> succinic anhydride / ethylene oxide copolymer X, and the biodegradability polyester copolymer was obtained like the example 1 except having made the catalyst addition into the 0.2 weight section to the mixture 100 weight section, using a succinic anhydride / ethylene oxide copolymer Z.

[0043] The biodegradability polyester copolymer was obtained like the example 9 except having used the mixture which made the <example 10> polyethylene terephthalate A, and the succinic anhydride / ethylene oxide copolymer Z mix by the weight ratios 70/30.

[0044] The biodegradability polyester copolymer was obtained like the example 1 except having made the catalyst addition into the 0.1 weight section to the mixture 100 weight section, using the mixture which made the <example 11> polyethylene terephthalate A, and the succinic anhydride / ethylene oxide

copolymer V mix by the weight ratios 70/30.
[0045] The biodegradability polyester copolymer was obtained like the example 1 except having made the catalyst addition into the 0.1 weight section to the mixture 100 weight section, using the mixture which made the <example 12> polyethylene terephthalate A, and the succinic anhydride / ethylene oxide copolymer W mix by the weight ratios 60/40.
[0046] The biodegradability polyester copolymer was obtained like the example 12 except having used the mixture which made the <example 13> polyethylene terephthalate A, and the succinic anhydride / ethylene oxide copolymer W mix by the weight ratios 80/20.
[0047] the mixture which made the <example 14> polyethylene terephthalate A, and the succinic anhydride / ethylene oxide copolymer X mix by the weight ratios 80/20 -- a high-speed high kneading biaxial extruder (the product made from an incorporated company techno bell "KZW15-60MG" --) 15mm and screw length (L) 900mm, [the diameter of a screw (D)] After ratio-of-length-to-diameter=60 and the vent section put in and carried out melting to the composition prepared four places, After having introduced the tetramer object of titanium (IV) tetrapod isopropoxide in the extruder so that it might become the 0.5 weight section to the aforementioned mixture 100 weight section, and kneading under reduced pressure conditions for 290-degree-C 30 minutes by screw-speed 400 revolution per minute, extrusion molding was carried out and the cylindrical Plastic solid was obtained. In addition, the moldability in the case of this extrusion molding (processability) was very good.
[0048] It replaced with the <example 1 of comparison> succinic anhydride / ethylene oxide copolymer X, and the biodegradability polyester copolymer was obtained like the example 1 except having used polybutylene succinate (succinic-anhydride content %, number average molecular weight 42000 of 50 mols).
[0049] Each biodegradability polyester copolymer obtained as mentioned above is meltable with chloroform, and performed characterization of a resultant by GPC, 1 H-NMR, and DSC. These results are shown in Tables 1 and 2. In addition, it also sets from the result of 1 H-NMR (solvent : CDCl3) to any of examples 1-14. It adds to the signal of the polyethylene-terephthalate origin, and the signal of a succinic anhydride / ethylene oxide copolymer origin. The methylene proton which shows combination of both the polymer unit (delta= 4.53 ppm) A delta= 4.43 ppm signal is seen and it could check that the obtained polymer was a polyester copolymer which the ester exchange reaction was made and was obtained between the polyethylene terephthalate, and the anhydrous dicarboxylic acid / ethylene oxide copolymer. Moreover, the number average molecular weight in Tables 1 and 2 was calculated by GPC measurement (standard sample polystyrene, solvent chloroform).
[0050] In addition, when the film was created by the solvent cast method, in the example 1 of comparison, it was translucent to being transparent in the examples 1-14.
[0051]
[Table 1]

	SA-EOの組成比（モル%） SA／EO	仕込重量比 PET/ SA-EO	収率 (%)	生成ポリエステル共重合体	
				組成（モル%） PET/ SA-EO	数平均分子量(Mn)
実施例1	49／51	50／50	88	74／26	38300
実施例2	49／51	50／50	72	68／32	28000
実施例3	49／51	70／30	62	84／16	32900
実施例4	49／51	60／40	72	68／32	31000
実施例5	49／51	80／20	70	76／24	27000
実施例6	49／51	40／60	72	56／43	37000
実施例7	49／51	20／80	70	24／76	25000
実施例8	47／53	50／50	70	68／32	26500

PET：ポリエチレンテレフタレート　　SA-EO：無水コハク酸／エチレンオキシド共重合体
収率：クロロホルム可溶部での収率

[0052]
[Table 2]

	SA-EOの組成比（モル%） SA／EO	仕込重量比 PET/ SA-EO	収率 (%)	生成ポリエステル共重合体	
				組成（モル%） PET/ SA-EO	数平均分子量(Mn)
実施例9	45／55	50／50	68	70／30	31000
実施例10	45／55	70／30	69	49／51	32000
実施例11	40／60	70／30	60	51／49	25000
実施例12	21／79	60／40	45	42／58	24000
実施例13	21／79	80／20	35	45／55	21000
実施例14	49／51	80／20	−	63／37	20000

PET：ポリエチレンテレフタレート　　SA-EO：無水コハク酸／エチレンオキシド共重合体
収率：クロロホルム可溶部での収率

[0053] Based on the following examining method, it evaluated about each above-mentioned biodegradability polyester copolymer.
[0054] The <biodegradability examining method> enzymatic hydrolysis examining method estimated biodegradability. namely, the enzyme after being immersed in pH 7.0 phosphate buffer solution (2mL) for 24 hours and keeping the sample (25mg) of each polyester copolymer at 37 degrees C -- the specified quantity -- in addition, it put for 24 hours, next, this liquid was filtered with the filter of 0.2 micrometers of apertures, the fixed quantity of the aqueous nature solution product was carried out using

the TOC measuring device (the "TOC10B type" by Shimadzu Corp.) about the filtrate, and this fixed quantity value was made into the index of 1250U As an enzyme, BERINGA Mannheim Rhizopus-stolonifer lipase (Rhizopus arrhizus 50000 U/mL) was used. In addition, the TOC value measured by the system in which an enzyme does not exist in the above and these conditions, and the system of only an enzyme was deducted as a blank value.

[0055] The film of each copolymer was created by the <viscoelastic-property examining method> solvent cast method, it was attached to this film, and viscoelastic property (tandelta, storage modulus) was measured using viscoelasticity spectrum meter by part for 10Hz [of oscillation frequency], and 2 degrees-C/of programming rates. In addition, it is known that the peak temperature of tandelta shows Tg (glass transition temperature) of polymer.

[0056] The piece for an examination of a dumbbell (refer to drawing 1) was created for the biodegradability polyester copolymer film created by the <test-for-tensile-strength method> solvent cast method using the dumbbell punching fixture, the tension test (test period : 200mm / per minute) was performed using this piece of dumbbell per Instron universal-testing-machine 4501 type, and tensile strength was measured.

[0057] The piece for an examination of a dumbbell (refer to drawing 1) was created for the biodegradability polyester copolymer film created by the <pace-of-expansion examining method> solvent cast method using the dumbbell punching fixture, and two indexes were carried out with the point in 20mm of mutual distance. The tension test (test period : 200mm / per minute) was performed using this piece of dumbbell per Instron universal-testing-machine 4501 type, the distance between indexes at the time of cutting was measured, and the pace of expansion was computed from the following formula.

[0058] Pace-of-expansion (%) ={(distance -20 between indexes at time of cutting)/20} x100[table 3]

	Ｔｇ (℃)	貯蔵弾性率Ｅ′ (ＭＰａ) (測定条件２０℃)	引張強度 (Kg/cm²)	伸び率 (%)	生分解性試験結果 水溶性有機炭素量の総量(ＴＯＣ) (ppm)
実施例１	３７	１．６	２２０	３７０	４０
実施例２	３０	０．６	１５０	２３０	３０
実施例３	４８	４．６	２８０	３０	２０
実施例４	３５	１．５	２１０	２００	２０
実施例５		２．０	２６０	３０	１５
実施例６	－	０．４	１２０	３６０	１２０
実施例７	---	０．０６	６０	２５０	２４００
実施例８	---	１．３	２６０	２０	３０

Ｔｇ：ｔａｎδのピーク温度より求めたもの

[0059]
[Table 4]

	貯蔵弾性率E′ (MPa) (測定条件20℃)	引張強度 (Kg/cm²)	伸び率 (%)	生分解性試験結果 水溶性有機炭素量の総量(TOC) (ppm)
実施例9	1．7	270	20	30
実施例10	0．06	70	260	140
実施例11	0．06	60	260	100
実施例12	0．04	50	280	240
実施例13	0．02	50	250	230
実施例14	1．3	230	50	20
比較例1	0．08	130	10	10

[0060] The polyester copolymer of the examples 1-14 of this invention could check sufficient storage modulus and sufficient tensile strength being obtained, and also providing the good elongation property good [intensity] while showing the outstanding biodegradability so that clearly from Tables 3 and 4. On the other hand, elongation with the sufficient polyester copolymer of the example 1 of comparison which does not have an ether chain was not acquired.

[0061]

[Effect of the Invention] It is excellent also in processability and its properties, such as intensity and shock resistance, are also good while it has the outstanding biodegradability, since the biodegradability polyester copolymer of this invention is a polyester copolymer obtained by performing an ester exchange reaction between a polyethylene terephthalate, and the anhydrous dicarboxylic acid / ethylene oxide copolymer which has an ethylene oxide content in the range below 100 mol % exceeding 50-mol %. There is an advantage which can improve biodegradability notably by having especially the ether chain of ethylene oxide. and rubber since it has the ether chain of ethylene oxide -- a character [like] is presented and sufficient elongation is also provided Therefore, in various uses, it can use suitably as a film, a sheet object, a Plastic solid, etc.

[0062] When an anhydrous dicarboxylic acid / ethylene oxide copolymer is the succinic anhydride / ethylene oxide copolymer expressed with the aforementioned general formula (I), while being able to improve further, intensity can also raise biodegradability further.

[0063] The more excellent biodegradability is securable, securing sufficient elongation, when the ethylene oxide content in an anhydrous dicarboxylic acid / ethylene oxide copolymer is 51-80-mol the range which is %.

[0064] More excellent intensity and elongation are securable, fully securing the outstanding biodegradability to the polyethylene-terephthalate 100 weight section, when the mixed weight ratio in the case of an ester exchange reaction is the range of an anhydrous dicarboxylic acid / ethylene oxide copolymer 20 - the 800 weight sections.

[0065] Moreover, according to the manufacture method of the biodegradability polyester copolymer concerning this invention, since generation of the copolymer by the ester exchange reaction and fabrication of this copolymer can be performed continuously, it is efficient and is suitable as a industrial process. Moreover, since it kneads by torque with a strong screw, an ester exchange reaction can fully be advanced for a short time. Furthermore, there is no dispersion in the quality of the biodegradability polyester copolymer obtained, and it excels in manufacture stability.

[0066] When an extruder is a biaxial extruder, the biodegradability polyester copolymer of the quality

stabilized more can be manufactured.
[0067] When the screw speed of an extruder is 150 - 600 revolution per minute, the biodegradability polyester copolymer no dispersion is [copolymer] in quality can be manufactured more efficiently in a short time.

[Translation done.]